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Capturing the complex physics behind universal grain size distributions in thin metallic films

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Abstract

Grain growth experiments on thin metallic films have shown the geometric and topological characteristics of the grain structure to be universal and independent of many experimental conditions. The universal size distribution, however, is found to differ both qualitatively and quantitatively from classical curvature driven models of Mullins type, which reduce grain growth to an evolution of a grain boundary network, with the experiments exhibiting an excess of small grains (termed an "ear") and an excess of very large grains (termed a "tail") compared with the models. While a plethora of extensions of the original Mullins model have been proposed to explain these characteristics, none have been successful. In this work, large-scale simulations of a model that resolves the atomic scale on diffusive time scales, the phase field crystal model, are used to examine the complex phenomena of grain growth. The results are in remarkable agreement with the prior experimental results, recovering the characteristic "ear" and "tail" features of the experimental grain size distribution. The simulations also indicate that, while the geometric and topological characteristics are universal, the dynamic growth exponent is not.

Keywords: Grain growth; Size distribution; Phase field crystal; Scaling law

1. Introduction

Most metals, ceramics and minerals are polycrystalline materials containing grains of different crystal orientation. The size, shapes and arrangements of these grains strongly affect macroscale material properties, such as fracture, yield stress, coercivity and conductivity. In magnetic systems, for example, the coercivity (or magnetic "hardness") can change by four or five orders of magnitude with a change in grain size [1]. Thus, understanding and controlling polycrystalline structures is of great importance in the production of many engineering materials, and has motivated numerous experimental and theoretical studies of grain growth.

Grain growth in thin metallic films is one example where extensive research has been conducted. One very interesting experimental finding in such systems is that the grain size distributions and topological characteristics appear to be independent of many experimental conditions [2]. More specifically, it has been found that, for a large collection of Al and Cu thin films, a universal grain size distribution emerges that is independent of the substrate, annealing temperature, purity, thickness and annealing time. Unfortunately the universal distribution is qualitatively and quantitatively different from the results of extensive computational studies on grain growth (e.g. [3]), which are based on the original Mullins model [4]. In this model, the problem is reduced to the evolution of a two-dimensional grain boundary network by relating the normal velocity v_n to the curvature κ of the grain boundary, $v_n = \mu\gamma\kappa$, with mobility μ and surface tension γ, and

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specifying the Herring condition [5] at triple junctions. Various attempts have been made to extend the original Mullins model and to include more realistic effects, such as interactions of the film with the substrate, anisotropy in the grain boundary energy and mobility, grain boundary grooving, and solute and triple junction drag (see Ref. [2] and the references therein). While these extensions have in some cases been shown to significantly alter the grain statistics, no single cause has been able to explain all of the experimental measured quantities, as discussed in Ref. [2]. This discrepancy raises the question of whether the underlying picture of an evolving smooth grain boundary network of the Mullins curvature-driven models is perhaps oversimplified.

In addition to the grain size distribution, the rate of growth of the average grain size has also been examined in detail. The original Mullins model and its extensions all seem to predict that the average grain size, represented by its radius $r(t)$, has a power law behavior of the form $\sim t^{1/2}$, which follows immediately from the linear relationship between grain boundary velocity and curvature. Experimentally, a much slower coarsening or even stagnation of grain growth in thin films is observed. This may be because the original Mullins model and its extensions ignore the crystalline structure of the grains, the dissipation due to lattice deformations and the Peierls barriers for dislocation motion. It is difficult to reconcile Mullins-type models with the atomistic features of grain boundaries, which (for low angles) can be seen as an alignment of dislocations, where the driving force for grain growth is the stress associated with dislocation motion. The differences of the description are shown schematically in Fig. 1.

2. Atomic considerations

Atomistic descriptions can incorporate the important physical features missing in the Mullins-type models and have led to some important observations. It has been shown that the complex dislocation structure along curved grain boundaries gives rise to a misorientation-dependent mobility [6]. Further studies indicate that grain boundaries undergo thermal roughening associated with an abrupt mobility change, leading to smooth (fast) and rough (slow) boundaries [7], which can eventually lead to stagnation of the growth process. The defect structure at triple junctions can lead to a sufficiently small mobility limiting the rate of grain boundary migration [8,9]. Also, tangential motion of the lattices is possible. For low-angle grain boundaries, normal and tangential motion are strongly coupled as a result of the geometric constraint that the lattices of two crystals change continuously across the interface while the grain boundary moves [10]. As a consequence of this coupling, grains rotate as they shrink, which leads to an increase in the grain boundary energy per unit length, although the overall energy decreases since the size of the boundary decreases [11–13]. Each of these phenomena can be simulated using molecular dynamics (MD) (see Ref. [14] for a review). However, to study the effect of these phenomena on scaling laws, grain size distributions or stagnation of growth requires a method which operates on diffusive time scales. For this reason, we choose to study the PFC model, which incorporates atomistic details on diffusive time scales.

3. PFC model

The PFC method [15] was introduced to model elasticity, dislocations and grain boundaries in polycrystalline systems in a simple and natural fashion. The model has been shown to successfully model grain boundary energies as a function of misorientation [16] and non-classical grain rotation during grain shrinkage and drag of triple junctions [17]. In addition, lower coarsening exponents have been observed for hexagonal lattices [18–20], and even stagnation of grain growth could be seen [21]. The aim of this paper is to use the PFC model on large scales to obtain statistical data for grain size distributions and to compare them with prior experimental data for thin metallic films. Since the experimental results in Ref. [2] seem to be universal, we do not fit the PFC parameters to a specific material

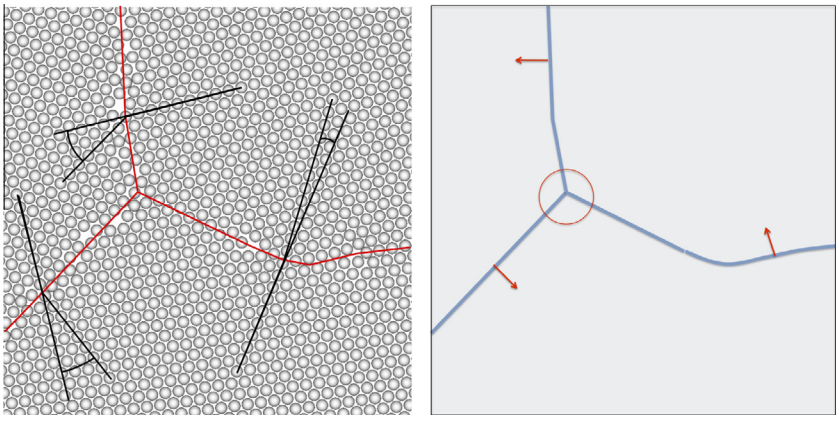

Fig. 1. Schematic comparison between an atomistic description of a polycrystalline material and a coarse-grained picture of a smooth grain boundary network. Shown is a low-angle grain boundary with aligned dislocations and two high-angle grain boundaries in an otherwise hexagonal lattice.